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CRITERIA FOR THE CERTIFICATION OF NON-RADIOACTIVE HAZARDOUS WASTE.

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ABSTRACT

In 1991, in response to the Department of Energy (DOE) Moratorium on the shipment of hazardous waste from Radioactive Materials Management Areas (RMMAs), Lawrence Livermore National Laboratory (LLNL) developed a process to use a combination of generator knowledge and/or sampling and analyses to certify waste as non-radioactive. The analytical process used the minimum detectable activity (MDA) as the de minimus value.

In the past twelve years, a great deal of operating experience has shown the LLNL certification process has serious limitations including:

- Procedure-specified analytical methodologies have resulted in the inability to adopt new techniques and methods that are more rapid, safer, and produce less waste.
- The characterization of materials as radioactive or non-radioactive is dependent on method-specific detection limits, not on an objective risk-based standard.
- There are substantial differences in the limits for surface contamination, sewer discharges, and hazardous waste moratorium determinations, even though all of these methods are used to free-release materials from radiological controls.

LLNL, in conjunction with the Chamberlain Group and Dade Moeller & Associates, Inc., is pursuing a risk-based approach to determine whether waste is non-radioactive, consistent with DOE guidance. This paper discusses the approach, which includes defining the radionuclides considered, establishing the exposure scenarios for the critical groups identified for each of three waste streams, defining the exposure pathways and key input data or assumptions, presenting radiation doses for unit concentrations of radionuclides in each waste stream, presenting radiation doses for unit concentrations of radionuclides in each waste stream, presenting the authorized limits for each waste stream, and discussing the results. Analytical values which fall below these authorization limits will be considered non-radioactive, with any individual dose maintained below 1 mrem/yr.

INTRODUCTION

On May 17, 1991, the Department of Energy (DOE) imposed a moratorium on the shipment of hazardous wastes to non-DOE Treatment, Storage, and Disposal Facilities (TSDFs) unless the generating facility could certify that the waste contained no "DOE Added Radioactivity." Each DOE facility was to develop site-specific procedures and analytical methodologies to meet this somewhat ambiguous criterion. Late in 1991, Lawrence Livermore National Laboratory (LLNL) developed a process to use a combination of generator knowledge and/or sampling and analyses to certify waste as non-radioactive. Generator knowledge was used as a first step to certify a waste as either definitely "rad added" and to be managed as radioactive, or definitely "non-rad added" and managed as non-radioactive. In cases where the waste generator could not determine whether the waste stream contained radioactive contamination, radioanalytical methods were used to make the determination. The methods chosen were liquid scintillation counting for tritium and gas-proportional counting for gross alpha/beta activity. The analytical process used the method and sample-size-specific minimum detectable activity (MDA) as the de minimus value. If the data showed activity above the MDA, the waste was determined to be radioactive. If the data showed no activity statistically above the MDA, the waste was determined to have no radioactivity added.

As technology has developed, the MDAs have been lowered; however, waste matrices tend to interfere with the MDAs, making it difficult to determine whether there is really radioactivity in the sample, or whether it is matrix interference. In addition, it is desirable to have limits for a “rad/non-rad” cutoff be independent of the method used to measure activity.

This paper contains technical information supporting the derivation of authorized limits, consistent with the guidance provided by U.S. Department of Energy (DOE) Order 5400.5 (1993), for one specific waste stream from the Lawrence Livermore National Laboratory (LLNL). In a memorandum dated November 17, 1995 (DOE 1995), DOE EH-412 provided clarification of issues related to DOE Order 5400.5 and the requirements for control of residual radioactive material.

EXPERIMENTAL

Wastes received at LLNL were divided into separate waste streams. The three specific waste streams considered in this overall analysis include: 1) non-sewerable aqueous liquids to be sent to a non-Nuclear Regulatory Commission licensed facility, 2) solids sent to a sanitary landfill, and 3) solids sent to a hazardous (class 3) landfill. The analysis of solid waste sent to a hazardous landfill is provided in detail for the purposes of discussion.

The goal of this effort was to produce authorized limits, in units of _Ci/mL for liquids and pCi/g for solids, for the three LLNL waste streams that would result in radiation doses less than 1 mrem/y to the most exposed individual in any realistic disposal scenario.

It is recognized that there is a good deal of uncertainty associated with the activities of individual members of the identified critical groups that may encounter the LLNL waste streams, or even other potential groups not considered in detail in this analysis. To compensate for this uncertainty, the overall authorized limits identified in the analysis considered the smallest (most limiting) concentration for each radionuclide across the scenarios identified for individuals associated with the disposition of the waste stream, for each of the identified radionuclide groups. To further compensate for uncertainty, attempts have been made to use conservative, yet realistic, assumptions, such as shipping distances, so that conservative authorized limits for the waste stream would be produced. The resulting authorized limits provide assurance that it would be unlikely that any individual associated with the wastes would receive a dose in excess of the targeted 1 mrem/y, independent of the location of the specific treatment or disposal facility selected, and independent of the total quantity encountered in a given year (within the identified limitations). The authorized limit for any mixture of the radionuclides can be found using the sum of fractions[1] rule.

Although a broad spectrum of radionuclides, including activation products, fission products, transuranics, and uranium of various isotopic compositions are potentially present during research activities at LLNL, for analysis purposes it is essential to screen radionuclides with similar properties and similar scenario-specific radiation doses into representative groups. As a starting point for solid wastes, the four radionuclide groups for clearance identified in HPS/ANSI N13.12 (1999) were considered. These groups include high dose alpha emitters, high dose beta emitters, general beta emitters, and other (low-dose) beta emitters. The radionuclide groups were assigned in HPS/ANSI N13.12 using judgment considering the range of modeling results for the scenarios considered and accounting for the conservative nature of the scenario analysis that was conducted. Further, HPS/ANSI N13.12 provided guidance for radionuclides not shown in the radionuclide groups, indicating that: *a comparison of the effective dose factors, by exposure pathway, listed in Table A.1 of NCRP Report No. 123I (NCRP 1996) for the radionuclides in question and the radionuclides in the general groups above* [in the standard] *shall be performed and a determination of the proper group made, based on similarity of the factors* (HPS/ANSI 1999). Using these groupings, a limited set of radionuclides was identified to be considered in the analysis. In recognition of potentially different behavior by radionuclides in the HPS/ANSI N13.12 groups as

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[1] A determination of whether or not a radionuclide mixture meets the authorized limits described in this report is made if the sum, over all radionuclides in the mixture, of the measured concentration of each

applied to the pathways and scenarios considered in the analysis, several radionuclides per group were included. Table 1 shows the HPS/ANSI N13.12 groupings.

Table 1. Radionuclides Considered by HPS/ANSI Group

HPS/ANSI N13.12 Group 1			**HPS/ANSI N13.12 Group 2**		
Radionuclide	Half-Life (y)	HPS/ANSI N13.12 Screening Level (pCi/g)	Radionuclide	Half-Life (y)	HPS/ANSI N13.12 Screening Level (pCi/g)
^{210}Pb +D	22	3	^{22}Na	2.6	30
^{226}Ra +D	1,600	3	^{60}Co	5.3	30
^{228}Ra +D	6.7	3	^{65}Zn	0.67	30
^{228}Th +D	1.9	3	^{90}Sr +D	28	30
^{230}Th	80,000	3	^{94}Nb	20,000	30
^{232}Th	1.4E+10	3	^{106}Ru +D	1	30
^{237}Np +D	2.1E+6	3	^{134}Cs	2.0	30
^{238}Pu	86	3	^{137}Cs +D	30	30
^{239}Pu	24,000	3	^{152}Eu	13	30
^{240}Pu	6,600	3	^{154}Eu	16	30
^{241}Am	460	3	^{234}U	25,000	30
^{244}Cm	18	3	^{235}U+D	7.1E+8	30
			^{238}U+D	4.5E+9	30

HPS/ANSI N13.12 Group 3			**HPS/ANSI N13.12 Group 4**		
Radionuclide	Half-Life (y)	HPS/ANSI N13.12 Screening Level (pCi/g)	Radionuclide	Half-Life (y)	HPS/ANSI N13.12 Screening Level (pCi/g)
^{36}Cl	31,000	300	^{3}H	12	3,000
^{129}I	1.7E+7	300	^{14}C	5,700	3,000
^{241}Pu +D	13	300	^{55}Fe	2.9	3,000
			^{63}Ni	92	3,000
			^{99}Tc	210,000	3,000

Finally, although a broad spectrum of radionuclides may be encountered at LLNL, some types of radionuclides are more likely to be present than others. For example, most fission products, including ^{36}Cl, ^{99}Tc, ^{129}I, are rarely encountered.

As the first step in the technical development of authorized limits, the likely disposition of each of the three waste streams was identified. In each case, the potential critical groups and maximally exposed individuals were considered. These individuals included

radionuclide divided by its authorized limit is less than or equal to one.

those that may come into contact with or may be located in the vicinity of the waste streams during transportation, treatment, and disposal. A unit radioactive concentration of 1 pCi/g of solid waste in a single waste shipment was assumed so that unit concentration dose conversion factors for each identified radiation exposure scenario and for each waste shipment and waste stream could be produced. These factors were developed to permit spreadsheet analyses and sensitivity studies to account for the predicted volumes of waste to be disposed in a given year in the development of the final authorized limits. The following paragraphs provide descriptions of the exposure pathways and scenarios associated with solid waste disposed of at a hazardous waste landfill.

For solid waste disposed of at a hazardous waste landfill, there were six major activities identified. The radiation dose calculations were conducted using the TSD-Dose (Pfingston et al. 1998) computer code. The waste was assumed to be transported in a transport truck, containing 20 tons (18 metric tonnes) of waste in about 200 drums. The TSD-Dose default large truck, with dimensions of about 16 m by 2.5 m by 2.8 m, was assumed. The hazardous waste landfill was conservatively assumed to be located 1,000 km (615 miles) away. It is noted that most hazardous waste facilities provide some type of respiratory protection for facility workers. Because the hazardous waste landfill is assumed to be a highly regulated facility, the post-closure scenarios for disposal of wastes at a sanitary landfill were considered to provide conservative upper bounds to the potential individual doses on a unit concentration, single shipment basis. The key TSD-Dose parameters required for this analysis are summarized in Table 2.

The following paragraphs briefly summarize the scenarios included in the analysis of LLNL solid wastes sent to a hazardous waste landfill.

Table 2. TSD-Dose Parameters for Solid Wastes Disposed at a Hazardous Waste Landfill

TSD-Dose Operation/Task	Worker	Distance From Waste (m)	Duration of Exposure (hours)	Shielding Thickness (cm)
Loading Operator	Load / Secure	0.91	4	0.32
Weight Station	Weigh / Inspect manifest	1.52	0.0833	0
Driver to Landfill	Driver			
	Load / Secure	0.91	0.167	0.32
	Drive	2.13	10	0.32
	Rest	0.61	1	0.32
	Maintenance	0.91	0.10	0.32
Waste Receipt	Detarping			
	Inspect / Sample	0.15	0.167	0.32
Tipping into Landfill		1.52	0.167	0.32
Cover Waste	CAT Operator	0.91	0.25	0.64

- Loading Operator. This activity involves loading and securing the waste in an open truck for transport to the hazardous waste landfill. External exposure is the only identified pathway using the parameters defined in Table 2.
- Weight Station. This activity includes weighing and inspecting the shipment against the manifest. External exposure is evaluated using the parameters defined in Table 2.
- Transport to the Hazardous Waste Landfill. Transporting the waste to the hazardous waste treatment facility involves the truck driver and consists of four activities: observing the waste loading operation, driving to the facility, rest en-route (assumed to occur in the back of the truck cab), and minor truck maintenance (checking the tires and refueling). External exposure is the only identified pathway using the parameters defined in Table 2.

- Waste Receipt/Detarping/Sampling. Upon arrival at the hazardous waste landfill, it is assumed that the tarp is removed from the waste and it is briefly inspected and weighed. External exposure and inhalation are the only identified pathways using the external exposure parameters defined in Table 2 and the TSD-Dose default inhalation parameters.
- Tipping Waste into the Hazardous Waste Landfill. After receipt at the hazardous waste landfill, it is assumed to be tipped (dumped) in the waste pit. The external exposure conditions and TSD-Dose parameters are defined in Table 2.
- Covering the Waste. Current hazardous waste landfill practices include covering the waste with clean dirt at least daily. This scenario describes the external exposure conditions of a heavy equipment (CAT) operator while covering the waste. The TSD-Dose parameters are defined in Table 2.
- Post-Closure Scenarios. The individual doses estimated for disposal of sanitary wastes for the ground water protection, park visitor, and human intrusion scenarios are assumed to provide conservative upper bounds for the impacts associated with disposal of wastes at a hazardous landfill.

As the first step in developing authorized limits for the LLNL waste stream, unit concentration radiation dose conversion factors for each of the identified radiation exposure scenarios and radionuclides were developed. These factors were generally based on a unit concentration (1 pCi/g of solid waste) of each reference radionuclide in each waste stream, for a single shipment of each type of waste.

For solid waste disposed of at a hazardous waste facility, the radiation doses estimated using TSD-Dose for the identified radiation exposure scenarios associated with disposal of a single shipment of LLNL waste were calculated in units of mrem/y per pCi/g per shipment. As could be expected, the detarping station scenario generally dominates the individual doses for several Group 1 radionuclides because inhalation exposures are assumed to occur.

The next step was to provide a derivation and an overall summary of the authorized limits for the waste stream. The general procedure used was to determine the inverse of the limiting scenario single shipment unit dose conversion factors, by radionuclide, with a determination of the radionuclide concentrations that would be protective of 1 mrem/y (i.e., pCi/g per 1 mrem/y for solids). As previously discussed, the goal of this analysis was to produce authorized limits that would result in radiation doses less than 1 mrem/y in a defendable and cost-effective manner, with consideration of administrative and practicality issues. For all of the waste categories considered, it was recognized that it is difficult to predict the total quantity of waste to be handled in any year, and that basing the authorized limit on the quantity of waste in one shipment would be quite limiting. The proposed authorized limits were developed considering the single shipment values, appropriate radionuclide groups and contamination levels protective of 1 mrem/y, consistent in instrumentation detection capabilities.

For operational effectiveness, it was determined that reducing the authorized limits for solid wastes to be consistent with the HPS/ANSI N13.12 screening levels, would be protective of 1 mrem/y, detectable using current practices, and allow for multiple shipments of waste per year.

The final LLNL authorized limits for solid wastes sent to a hazardous waste facility are the clearance levels from HPS/ANSI N13.12 in units of pCi/g. The maximum number of shipments per year was determined, at the final authorized limit, to equal 1 mrem/y, assuming that the same individuals are exposed for each scenario, for all shipments. Up to 43 shipments per year of solid wastes could be shipped to a hazardous waste landfill with an authorized limit of 30 pCi/g for Group 2 radionuclides, without exceeding 1 mrem/y. Since each shipment was assumed to contain 20 tons (18 metric tonnes), the total shipped per year should not exceed 850 tons (770 metric tonnes).

RESULTS

An overall summary of the authorized limits requested for the waste stream is provided in Table 3. Solid waste authorized limits, in units of pCi/g, are included. The maximum quantities (in ton per year) of the waste stream, representing the limiting quantity for the limiting radionuclide group, is also included.

The authorized limit for any mixture of the reference radionuclides, or groups of radionuclides, shown in Table 3 for the waste stream is found using the sum of fractions rule. A determination of whether or not a radionuclide mixture meets the authorized limits is made if the sum, over all radionuclides in the mixture, or all groups of radionuclides, of the measured concentrations divided by its authorized limit is less than or equal to one.

DISCUSSION

During the process of determining authorization limits, the approach was modified several times. The first approach was to use an extremely conservative risk-based model. Due to the variety of radionuclides used in a research and development facility, the decision was to assume the same people would be exposed to the waste every time a shipment was made to an off-site facility, and that the facilities were on the east coast.

Table 3. Overall Summary of Requested Authorized Limits

Waste Stream	**Radionuclide Group**	**Limiting Radionuclide**	**Final LLNL Solid Waste Authorized Limits: HPS/ANSI N13.12 Screening Group (pCi/g)**	**Maximum Quantity per year to be Disposed**
	HPS/ANSI N13.12			
	1	^{226}Ra +D	**3**	
Solid Waste Disposal	2	^{60}Co	**30**	
at a Hazardous Waste	3	^{241}Pu	**300**	
Facility	4	^{3}H	**3,000**	**850 (tons/y)**

(a) This value is significantly larger than the predicted annual generation rates for the waste stream shown. Because of the comparatively small quantity of waste to be disposed, further assurance is provided that individual doses will not exceed 1 mrem/y.

When the total volumes of waste per year were determined from this model, they were less than the current volumes encountered. Thus, some of the conservatism was removed from the model. LLNL researched the volumes that were necessary, and the models were re-worked to arrive at an acceptable level.

LLNL originally started with four waste streams in this model. The fourth waste stream, organic liquids, was treated in the model as aqueous waste. However, the authorization limits, especially for alpha radionuclides, was so low that they were not possible to detect in the laboratory given the waste oil matrix. This waste stream was removed from the model.

CONCLUSION

In conducting the analysis of authorized limits for the solid Lawrence Livermore National Laboratory waste streams, attempts were made to identify realistic end-uses associated with treatment and disposal of the wastes and to conduct a conservative analysis such that it would be unlikely that any individual would receive a dose in excess of 1 mrem/y. Some of the more important assumptions contributing to the conservative nature of the results included:

- Assuming uniform distributions of radioactive contaminants in the wastes since, in many cases, these wastes will be mixed with non-radioactive wastes from other (non-DOE) sources. Non-uniformity would change (likely increase) the assumed shielding properties associated with the external exposure pathway, the dominant exposure pathway for several of the reference radionuclides.
- Assuming that the quantity of waste released in a year would fill an entire truck load. Historical records for many of these wastes (with the possible exception of sanitary wastes and decommissioning rubble) would indicate that these volumes are conservatively large. Smaller quantities of wastes per shipment, and co-

shipment with wastes from non-radiological areas would reduce the doses to the maximally exposed individual proportionally.

- Assuming conservative, yet reasonable, transportation distances would bound the potential doses for shipment to other, more nearby, facilities.
- Deriving authorized limits based on the limiting scenarios and radionuclides would further assure that it would be unlikely that individual doses in excess of 1 mrem/y could be expected.
- Selection of HPS/ANSI N13.12 screening values, by radionuclide group, instead of the estimated authorized limits based on administrative and practicality issues produced conservative results.

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